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On the excitation energy of the ground state in the third minimum of ^{234}U

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Abstract

The ^{233}U(d,pf)^{234}U reaction has been studied with high energy resolution. The observed fission resonances in the excitation energy range of 4.75 $\leq E^* \leq$ 5.40 MeV were described as members of rotational bands with rotational parameters characteristic to the hyperdeformed nuclear shape ($\hbar^2/2\theta = 2.1 \pm 0.2$ keV). Information on the K values of the bands has been obtained from fission fragment angular distribution measurements. The level density of the most strongly excited $J = 3$ states has been compared to the prediction of the back-shifted Fermi-gas formula and the energy of the ground state in the third minimum has been estimated to be E_{III} = 3.1 $\pm$ 0.4 MeV. © 1999 Published by Elsevier Science B.V. All rights reserved.

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Recently, very effective, high resolution 4π gamma-ray spectrometers like EUROBALL and GAMMASPHERE have been developed for nuclear structure studies. The extensive development of these spectrometers was started about ten years ago after the discovery, in deformed nuclei, of high-spin superdeformed (SD) states [1,2] with a ratio of 2:1 for the long to the short axis. Today, one of the main goals of these spectrometers is to search for hyperdeformed (HD) nuclear shapes with an axis ratio of 3:1.

Early evidence for hyperdeformation in ^{152}Dy was reported by Galindo-Uribarri et al. [3]. Discrete transitions have been tentatively assigned to a HD band in ^{152}Dy by Viesti et al. [4]. LaFosse et al. [5] have made a more definite observation of a HD band in ^{147}Gd, but one year later they showed that the candidates previously reported did not have properties consistent with band structure [6].

In the actinide region a third minimum in the potential energy (which contains HD states) was predicted already more than twenty years ago by

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Möller et al. [7]. According to recent calculations, in these nuclei the so-called third minimum of the potential barrier appears with deformation parameters $\beta_2 \approx 0.90$ and $\beta_3 \approx 0.35$ [8,9] and the depth is predicted to be much larger ($\Delta E \approx$ 3 MeV [10]) than believed earlier [11].

The excited states in the third minimum of several Th isotopes were investigated by Blons et al. [15] by measuring the micro-structure of sub-barrier fission resonances, but until now there is no experimental information available for the depth of the third minimum.

The γ and conversion-electron spectroscopy investigations of the SD states turned out to be very difficult in the actinide region because of the very low partial cross-sections (0.001% of the total), and the high background produced by the fission fragments. After the discovery of high-spin superdeformation in the $A = 150$ region, excited by a $\approx 5\%$ fraction of the total cross-section, the main focus of the research moved to that region although many interesting problems remained also in the actinide region.

One of the characteristic features of the HD bands in the actinide region, besides the large moment of inertia, is the appearance of octupole bands. Blons et al. [15] analyzed the micro-structure of the fission resonances in Th isotopes by assuming HD octupole-deformed rotational bands. These bands have been observed also in the first well in the actinide region (see e.g. [12]). The different consequences of the octupole deformation have been reviewed recently by Butler and Nazarewicz [13].

In our previous work [14] we reanalyzed the fission resonances in ^{234}U measured by Blons et al. [15] and showed that the unresolved peaks around $E^* = 4.9$ MeV could be interpreted as HD states in the third well of the potential barrier.

The aim of the present work is to study the ^{233}U(d,pf)^{234}U reaction with better energy resolution than Blons et al. [15], to resolve the HD rotational bands and, from the level densities, to estimate the depth of the third minimum.

In order to investigate the HD bands the excitation energy was chosen between the energy of the inner and outer barriers of the second well, i.e. between 4.5 and 5.2 MeV [14]. In this energy range the widths of the SD resonances in the second well should be much broader than those of the HD states due to the strong coupling to the normal deformed states. The widths of the HD states due to the higher outer barriers of the third well remain below the actual experimental resolution of ~ 5 keV.

The experiment on ^{234}U was carried out with a $E_d = 12.5$ MeV deuteron beam of the Munich Tandem accelerator. Enriched (99%) $\approx$ 30 μg/cm^2 thick targets of ^{233}U were used. The energy of the outgoing protons was analyzed by a Q3D magnetic spectrograph with a solid angle of 10 msr [16], which was set at $\Theta_{\rm Lab} = 130°$ relative to the incoming beam. The position of the analyzed particles in the focal plane was measured with a light-ion focal-plane detector of 1.8 m active length using two single-wire proportional counters surrounded by etched cathode foils [17]. A line-width of $\leq$ 3 keV has been observed for elastic scattering of 20 MeV deuterons. Fission fragments were detected by two position-sensitive avalanche detectors (PSAD) [18] having two wire planes (with delay-line read-out) corresponding to horizontal and vertical directions. Protons were measured in coincidence with fission fragments. The obtained proton–fission fragment coincidence spectrum is shown in Fig. 1. as a function of excitation energy. Recently we have performed a similar analysis also for ^{240}Pu where a much less complex (superdeformed) band structure was analyzed [19].

The obtained widths of the peaks show the experimental energy resolution up to about 5.3 MeV. Above this excitation energy the peaks get increasingly broader due to the increasing fission width when we

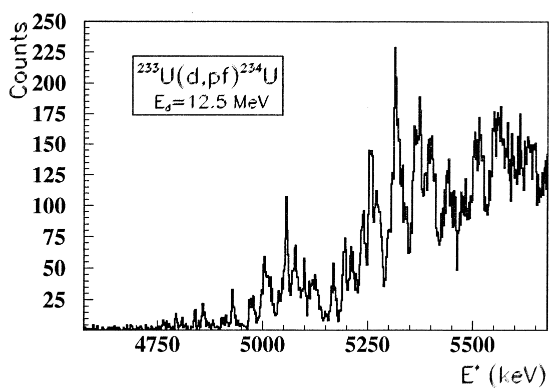

Fig. 1. Proton spectrum measured in coincidence with the fission fragments.

approach the top of the fission barrier (see Fig. 1 in Ref. [14]). The low energy part of the spectrum, which we analyzed before [14], is compared to the one published by Blons et al. [15] in Fig. 2.

Comparing the spectra we can conclude that the energy resolution has been considerably improved and we can clearly see the fine structure of the peaks. The energy calibration was taken from the ^{208}Pb(d,p) reaction, using the (d,p) Q-value of 1.710 $\pm$ 0.015 MeV [20]. According to this calibration the energy spectrum shown in the upper part of Fig. 2 had to be shifted by ≈ 150 keV from the one published by Blons et al. [15] to achieve consistency.

Experimentally the very large quadrupole and octupole moments of the HD states should manifest themselves by the presence of alternating parity bands with very large moments of inertia [15]. Assuming overlapping rotational bands with the same moment of inertia, inversion parameter [12] and intensity ratio for the members in a band, we fit our spectrum using simple Gaussians for describing the different band members in the same way as we did it in our previous work [14]. The result of the fit is shown in Fig. 3a).

The relative intensities of the members of the rotational bands have been taken from Back et al. [21] and are given in Table 1. These values have

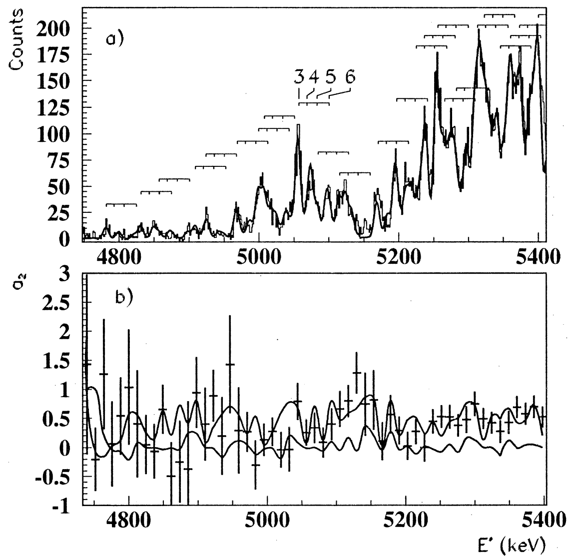

Fig. 3. (a) Part of the measured proton energy spectrum fitted with 24 rotational bands with a common rotational parameter. The spectrum was divided into two parts at $E = 5150$ keV for the fitting; (b) Experimental fission-fragment angular-distribution coefficients as a function of excitation energy compared to the calculated ones using $K = 1$ (upper curve) and $K = 3$ (lower curve) for all of the bands. The $K = 0$ curve is very close to the $K = 1$ one while the $K = 2$ is in between the $K = 1$ and $K = 3$ ones. They are not shown.

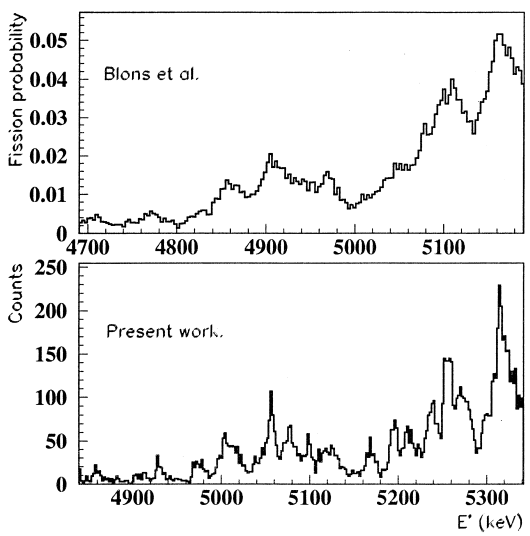

Fig. 2. Part of the proton spectrum measured in coincidence with the fission fragments and compared to the result of Blons et al. [15].

been calculated by DWBA for ^{235}U(d,p). These relative intensities depend sensitively on the distribution and width of the single particle states involved in the (d,p) process at the given excitation energy. We adopted these relative intensities as initial parameters for the fitting procedure. The final values are shown in Table 1.

The jump in the relative intensities going from $J = 2^+$ to 3^-, as predicted by Back et al. [21] might be a consequence of the alternating parities within the rotational bands, although our fit was actually not very sensitive to the relative intensity of the 2^+ state. Increasing this intensity by a factor of two worsens the χ^2 by less than 8%.

After fixing the relative intensities of the band members, we have used two specific parameters for each band during the fitting procedure: the energy of the band head and the absolute intensity of the band. A common rotational ($\hbar^2/2\theta$) and inversion splitting (ΔE_{+-}) parameter was adopted for each band.

Table 1
Relative intensity ratios of the rotational-band members populated in the ^{233}U(d,pf) reaction. The calculated values marked by a) and b) are taken from Ref. [20] for $\pi = +$ and $-$, respectively. The underlined values represent the relative intensities for an alternating parity, octupole rotational band. The adopted values were obtained from a fit of the 5.1 MeV region of the energy spectrum.

J	0	1	2	3	4	5	6	7	8
a)	<u>0.04</u>	0.04	<u>0.11</u>	0.50	<u>0.85</u>	0.69	<u>0.60</u>	0.50	<u>0.23</u>
b)	<u>0.08</u>	<u>0.23</u>	0.50	<u>1.00</u>	1.19	<u>1.00</u>	0.92	<u>0.23</u>	0.42
adopted	0.00	0.03	0.10	1.00	0.59	0.37	0.32	0.06	0.06
Ref. [23]	0.47	0.60	1.00	0.58	0.22	0.15	0.11	0.04	0.07

The result of the χ^2 analysis as a function of $\hbar^2/2\theta$ and ΔE_{+-} is shown in Fig. 4.

Although the statistics in the second part of the spectrum is better, the density of the states is about two times larger, making the determination of the rotational parameter more uncertain. The rotational parameter was therefore determined for both parts separately and the weighed average was calculated. As a result we obtained: $\hbar^2/2\theta = 2.2 \pm 0.2$ keV and $\Delta E_{+-} = 0^{+10}_{-15}$ keV.

The low-lying rotational bands have been investigated in the ^{233}U(d,pf) reaction at 13 MeV by Bjørnholm et al. [23]. They obtained intensity distributions, which peak around $J = 2$, distinctly lower than those of Back et al. (see Table 1). Although they investigated only quadrupole rotational bands with the same parity for the members of the band, and it is known that the intensities depend strongly on parity, we did use also their relative intensities to fit our data as an alternative scenario. Using alternatively at face value the relative intensities of Bjørnholm et al. [23] we obtained almost as a good fit as above with: $\hbar^2/2\theta = 1.5 \pm 0.5$ keV and $\Delta E_{+-} = 0 \pm 15$ keV. The somewhat different values obtained by assuming these two very different sets of relative intensities indicates the sensitivity of the rotational parameter for the assumed relative intensities. Although this dependence is of systematic nature, we quote here as the final value of the rotational parameter its weighed of the two analyses: $\hbar^2/2\theta = 2.1 \pm 0.2$ keV.

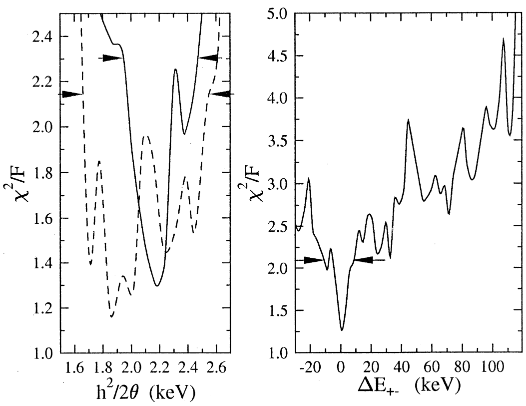

Fig. 4. The results of the χ^2 analysis for the first and second parts of the spectrum (full and dashed lines, respectively) as a function of the rotational ($\hbar^2/2\theta$) and inversion splitting (ΔE_{+-}) parameters, using the adopted relative intensities in Table 1.

As an a posteriori test one may write the relative intensities of the band members with different J_f as [24]:

$$R(J_f) = |\langle J_i, K_i, j, \Omega | J_f, K_f \rangle|^2 A(j), \tag{1}$$

with $J_i = K_i = 5/2$ for ^{233}U. The information on reaction mechanics and nuclear structure is entirely contained in the coefficients $A(j)$. If we assume $K_f = 0$, then $\Omega = 5/2$. Assuming only three contributing j transfers: $5/2$, $7/2$ and $9/2$ and using the $A(j)$ as free parameters we find that Bjørnholm's relative intensities are easily reproduced and the χ^2 minimum is found again at $\hbar^2/2\theta = 1.5$ keV and $\Delta E_{+-} = 0$ keV.

Allowing also higher j transfers and both $K_f = 0$ and 1, Back's relative intensities can be fairly well approximated. The χ^2 minimum is indeed around $\hbar^2/2\theta = 2.2$ keV, but is very shallow, due to the increased number of free parameters.

In both cases the obtained rotational parameters agree with the value corresponding to the HD shape and obtained in our previous work [14]. The $\Delta E_{+-} \approx 0$ value is consistent with the small inversion parameters obtained by Blons el al. [15] for the Th isotopes.

Although we made the fit very carefully we are aware of the fact that our analysis is not sensitive to the parity of the states. In case of $K \neq 0$ we can not distinguish between quadrupole and octupole rotational bands.

Assuming that we saw rotational bands built on some excited states, we tried to vary also the K value of the band head during the fitting procedure. However, the result of the fit was found to be insensitive to the K value when it was varied between 0 and 2, because the relative intensity of the members of the band with a $J \leq 2$ is much less compared to the intensity of the $J = 3$ line (see Table 1). In case of the quadrupole scenario we used $K = 1$ for all of the bands.

Fission-fragment angular distributions were generated as a function of the excitation energy, normalized to the known (d,f) angular distribution [25] and fitted with even Legendre polynomials (LP) up to fourth order. The a_2 angular distribution coefficient is shown in Fig. 3b as a function of the excitation energy. In order to get information on the spins and K values of the observed rotational bands, or to check our assumptions made for fitting the energy spectrum, the angular distribution coefficients of the fission fragments have been calculated and compared to the experimental ones.

In the Plane Wave Born Approximation the probability to detect a fission fragment at an angle θ relative to the classical recoil axis is [22]:

$$W_{K_f}^{J_f}(\theta) = \sum_{M_i, M_f, j, m} \frac{\sigma(j)}{\sigma} \frac{2j+1}{2(2J_f+1)} \left(C_{M_i m M_f}^{J_i j J_f} \right)^2 \times W_{M_f K_f}^{J_f}(\theta), \tag{2}$$

with

$$W_{MK}^{J}(\theta) = \tfrac{1}{4}(2J+1)\left(|D_{MK}^{J}(\theta)|^2 + |D_{MK}^{J}(\theta+\pi)|^2 \right),$$

for fission through a transition state with quantum numbers J_f, M_f, K_f, spin J_i, M_i of the target nucleus, and the total angular momentum j and m of the neutron transferred in the (d,p) reaction. The relative population of the states through different j transfers $(\sigma(j)/\sigma)$ has been taken from Ref. [23] but the angular distribution coefficients were actually not sensitive to this choice. All quantum numbers refer to the recoil axis. Expressing $W_{MK}^{J}(\theta)$ in terms of Legendre polynomials [26]:

$$W_{MK}^{J}(\theta) = \frac{2J+1}{2}(-)^{K-M} \sum_{\lambda}^{0,2,4,\ldots,2J} C_{-MM0}^{JJ\lambda} \times C_{-KK0}^{JJ\lambda} P_\lambda, \tag{3}$$

one gets the following expression for the angular distribution coefficients:

$$A_\lambda(J_f, K_f) = \sum_{M_i, M_f, j, m} \frac{\sigma(j)}{\sigma} \frac{2j+1}{2(2J_f+1)} \times \frac{2J_f+1}{2}(-)^{K_f-M_f}\left(C_{M_i m M_f}^{J_i j J_f} \right)^2 \times C_{-M_f M_f 0}^{J_f J_f \lambda} C_{-K_f K_f 0}^{J_f J_f \lambda}. \tag{4}$$

The $a_2(E^*)$ shown in Fig. 3b has been calculated by using the parameters obtained from a fit of the energy spectrum and by multiplying the amplitudes of the different band members by the corresponding $A_\lambda(J_f, K_f)$ values as well as by normalizing the whole distribution with the one calculated with the $A_0(J_f, K_f)$ values.

Since the spins J_f of the excited states are already fixed by the energy spacings of the peaks as shown in Fig. 3a in the next step only the K values of the bands were varied between 0 and 3. The measured angular distribution coefficients were compared to the calculated values in Fig. 3b.

In case of assuming quadrupole rotational bands with intensities peaking at $J^\pi = 2^+$ even the gross structure of the measured angular distribution coefficients could not be explained. The calculated a_2 coefficients were always too low compared to the experimental ones. This can be understood since $a_2 = -0.20$ for $J = 2$ and 0.03 for $J = 3$ and is increasing with J. In order to explain the experimentally measured $a_2 \approx 0.5$ values we should assume an intensity distribution which peaks at higher J values than 2, most probably at $J = 3$ as assumed before.

The density of the $J = 3$ states has been determined from our experimental data. The average distance of the two closest neighbors of a given state is shown in Fig. 5. The level spacing distribution is close to a Wigner distribution [27] but the mixing-in of some Poisson type distribution is also visible. The density of $J=3$ states has been calculated as a function of the excitation energy using the back-shifted Fermi-gas description with parameters determined by Rauscher et al. [28]. In order to estimate the depth of the third well we compared the experimentally obtained and calculated values. We assumed that the same parametrization of the level density formula is valid in the third well, as was determined by Rauscher et al. [28] by fitting the level densities in the first well of the potential barrier. This assumption is based on the finding of Glässel et al. [22] that the level density of the 2^+ states lying in the second well of ^{240}Pu is the same as the level density in the first well. The shell-correction energy used to determine the level density parameter ''a'' has been taken from the work of Möller et al. [29] while the spin cutoff-parameter σ was determined by using the rigid rotor rotational parameter suggested by Rauscher et al. [28]. The result of the comparison is shown in Fig. 5.

The calculated curve had to be shifted by 2.7 MeV to reproduce the experimental values. According to this comparison the ground state in the third well is shifted up by 2.7 MeV, which also means that the ''microscopic correction'' C(N,Z) in Eq. (14) of Ref. [28] should be modified by this energy and the level distances should be recalculated. Doing this in a recursive way we find a value of 3.1 MeV for the energy of the ground state in the third well (curves connected with an arrow ''Rauscher 1'' in Fig. 5).

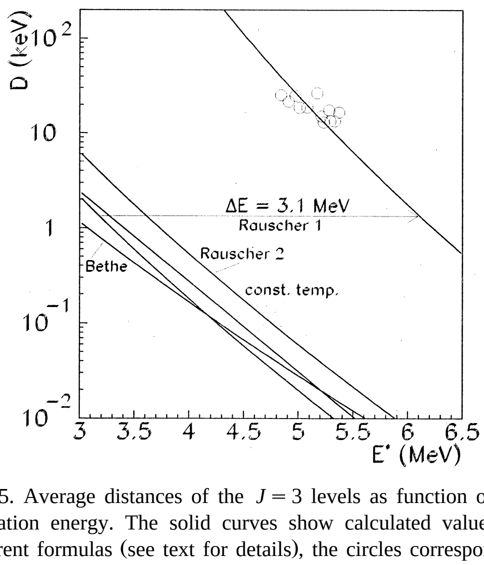

Fig. 5. Average distances of the $J=3$ levels as function of the excitation energy. The solid curves show calculated values by different formulas (see text for details), the circles correspond to experimental values.

Taking into consideration the shell and nucleon pairing correlation effects, Mughabghab and Dunford [30] calculated and fitted the spin cutoff parameter as a function of the atomic mass and found large deviations from the one obtained with the rigid rotor rotational parameter.

In order to get some estimate for the precision of the level distance analysis described above we repeated the calculation of level distances by using the rotational parameter deduced in the present work ($\hbar^2/2\theta = 2.1$ keV, ''Rauscher 2'' curve in Fig. 5). We also used two other formulas to estimate the level distances, which were parameterized by von Egidy et al. [31]. They used a constant temperature level density formula and the Bethe formula for the back-shifted Fermi gas model. The theoretical curves (also shown in Fig. 5) are calculated with these two formulas and parameters determined by von Egidy et al. [31] by fitting the low-lying level scheme ($E^* \leq$ 1.5 MeV) of ^{234}U. From the uncertainties of the calculated and measured level distances the error of the energy determination is estimated to be 0.4 MeV.

Ćwiok et al. [10] predicted two different HD minima for ^{234}U with very different β_λ ($\lambda = 3$–7) values. One of them has an octupole deformation parameter of $\beta_3 \approx 0.4$ and a minimum of $E_{\mathrm{III}} =$ 3.5 MeV while the other is more reflection-asymmetric and has an octupole deformation parameter of $\beta_3 \approx 0.6$ and a minimum of $E_{\mathrm{III}} = 2.7$ MeV. The experimental value of $E_{\mathrm{III}} = 3.1 \pm 0.4$ MeV obtained in the present work is between the two predicted values with an error bar, which overlaps both theoretical values. At this moment we do not have information on the β_3 of this nucleus.

In summary, we have measured the fission probability of ^{234}U as a function of excitation energy with high energy resolution using the (d,pf) reaction. The rotational parameter obtained from fitting the energy spectrum around $E^* \approx 5$ MeV is found to be $\hbar^2/2\theta = 2.1 \pm 0.2$ keV, which is characteristic for the hyperdeformed nuclear shape. The level density of the most strongly excited $J=3$ states has been compared to the prediction of the back-shifted

Fermi-gas formula and the energy of the ground state in the third minimum has been estimated to be $E_{\mathrm{III}} = 3.1 \pm 0.4$ MeV which agrees well with the predicted one [10].

Acknowledgements

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